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TECHNICAL REPORT

STUDY OF DETONATION BEHAVIOR
OF SOLID PROPELLANTS

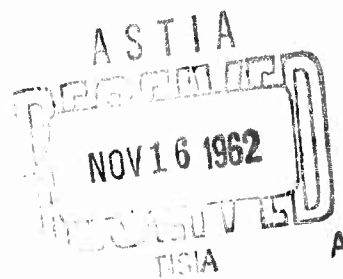
THIRD QUARTERLY REPORT

Prepared for: Department of the Navy
Bureau of Weapons
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SECTION 1

INTRODUCTION

This program is directed to a theoretical treatment of detonation behavior by numerical integration of the basic time dependent equations which define the detonation process. Ultimate objective is to perform such integration in two space dimensions so that the effect of geometry upon behavior (for example the phenomena of critical size) can be predicted in the case of large charge sizes. The detailed form of the basic equations, and numerical procedures¹ used in their integration have been described in previous reports.

During the present report period, one minor refinement has been made in the mathematical procedures. This was primarily in the interest of completeness, rather than any anticipated significant effect on the results of the integration. An important development has been the discovery that the use of the von Neuman "q" in integrating through a reactive shock can, under some circumstances, falsify the entropy increase and, as a consequence, seriously affect the computed final steady state propagation velocity. This is now regarded as the most likely explanation for the high computed steady state velocities discussed in the last quarterly² and previous reports³. Some changes in procedure will be required as a consequence of this discovery.

The two dimensional program is now in the final checkout and debugging stage. It is being actively pressed and should be completed soon. This will be affected somewhat by the problem associated with the von Neuman "q" described above.

The equation of state work is progressing at an accelerated pace. Data are presented in this report on polyurethane. Procurement of data on inorganic salts such as ammonium perchlorate, and other organic binders, and composite mixtures of the two is presently underway.

SECTION 2

ONE DIMENSIONAL METHODS

2.1 MATHEMATICAL DETAILS

The one dimensional computer program has been subjected to refinement in the detailed treatment of energy exchange between the solid and gaseous phases as discussed in Reference 3, Equations 2-6, inclusive. In this derivation, terms involving $d\gamma$ previously were neglected as a matter of convenience. The effect of this was regarded to be negligible; nevertheless, in the interest of completeness, modification of the program to include such terms has been made.

Starting with Equation 2 of Reference 3, one has*:

$$(p + \beta)v_s = (\gamma_s - 1)e_s + c$$

$$(p + \beta)dv + v_s dp = (\gamma_s - 1)de_s + e_s d\gamma_s$$

Since

$$de_s = -pdv_s$$

$$\left[\frac{\beta}{p} + \gamma_s - \frac{e_s}{p} \frac{d\gamma_s}{dv_s} \right] pdv_s = -v_s dp$$

$$\frac{d \ln p}{d \ln v_s} = - \left[\frac{\beta}{p} + \gamma_s - \frac{e_s}{p} \frac{d\gamma_s}{dv_s} \right] \quad 3(s)$$

*Symbols are defined in Reference 3.

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and for the gas ($\beta = 0$)

$$\frac{d \ln p}{d \ln v_g} = - \left[\gamma_g - \frac{e_g}{p} \frac{d \gamma_g}{d v_g} \right] \quad 4(s)$$

Equation 6, Reference 3, then becomes

$$dv_g' = \frac{d\bar{v} - (v_s - v_g) df_s}{1 - f_s \left[\frac{v_s \left(\gamma_g - \frac{e_g}{p} \frac{d \gamma_g}{d v_g} \right)}{v_g \left((\beta/p) + \gamma_s - \frac{e_s}{p} \frac{d \gamma_s}{d v_s} \right)} \right]} \quad 6(s)$$

With this change, Equations 20 and 21 become

$$v_s^{n+1} = v_s^n + \frac{(v^{n+1} - v^n) - (v_g^n - v_s^n) (f_s^n - f_s^{n+1})}{f_s^{n+1} + (1 - f_s^{n+1}) \left(\frac{v_g^n}{v_s} \right) \frac{\Gamma_s^n}{\Gamma_g^n}} \quad 20(s)$$

$$v_g^{n+1} = v_g^n + \frac{(v^{n+1} - v^n) - (v_g^n - v_s^n) (f_s^n - f_s^{n+1})}{1 - f_s \left[1 - \left(\frac{v_s}{v_g} \right) \frac{\Gamma_g^n}{\Gamma_s^n} \right]} + \frac{(\gamma_g^n - 1) \left[(e_s^n - e_g^n) (f_s^n - f_s^{n+1}) + \Omega^{n+1} \right]}{(1 - f_s^{n+1}) \left[p^n \gamma_g^n + (p^n \gamma_s^{n+\beta}) \left(\frac{v_g^n}{v_s} \right) \left(\frac{1 - f_s^{n+1}}{f_s^{n+1}} \right) \right]} \quad 21(s)$$

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where:

$$\Gamma_s^n = \gamma_s^n + \frac{\beta}{p^n} - \frac{e_s^n (\gamma_s^n - \gamma_s^{n-1})}{p^n (v_s^n - v_s^{n-1})}$$

$$\Gamma_g^n = \gamma_g^n - \frac{e_g^n (\gamma_g^n - \gamma_g^{n-1})}{p^n (v_g^n - v_g^{n-1})}$$

$$\Omega^{n+1} = F_2 Q_2 (f_2^n - f_2^{n+1}) + F_3 Q_3 (f_3^n - f_3^{n+1}) + Q_4 (f_4^n - f_4^{n+1})$$

The differencing scheme for evaluation of $\frac{d\gamma}{dv}$ is not of high accuracy. However, it is regarded as suitable for present purposes.

As expected, the effect of this correction on wave behavior is found to be negligible for the cases presently under treatment. This is shown by the wave trajectory of Figure 1, curve "a" which was computed using the corrected program and the same input data as for the trajectory shown on Figure 5 of Reference 1. The two curves are essentially identical.

2.2 STEADY STATE PROPAGATION VELOCITY

Recent one dimensional calculations on TNT have encountered difficulty in that the ideal steady state detonation velocities obtained with computed waves are about 12 mm./μ sec, 60 percent higher than the values predicted from steady state theory. The explanation for this discrepancy has been subjected to considerable scrutiny during this report period, since the correct prediction of steady state values is an important test of the validity of the computational techniques. As discussed briefly in Reference 2, one possible explanation for the high steady state velocity is that values used for the equation of state parameters are incorrect.

The sensitivity of final wave propagation velocity to one of the equation state parameters was examined by comparison of the trajectories obtained at two difference values of the parameter "b" which defines γ according to the expression

$$\gamma = \gamma_o + \left[\left(\frac{b}{v} \right) + \left(\frac{b}{v} \right)^2 \right] (\gamma_o - 1)$$

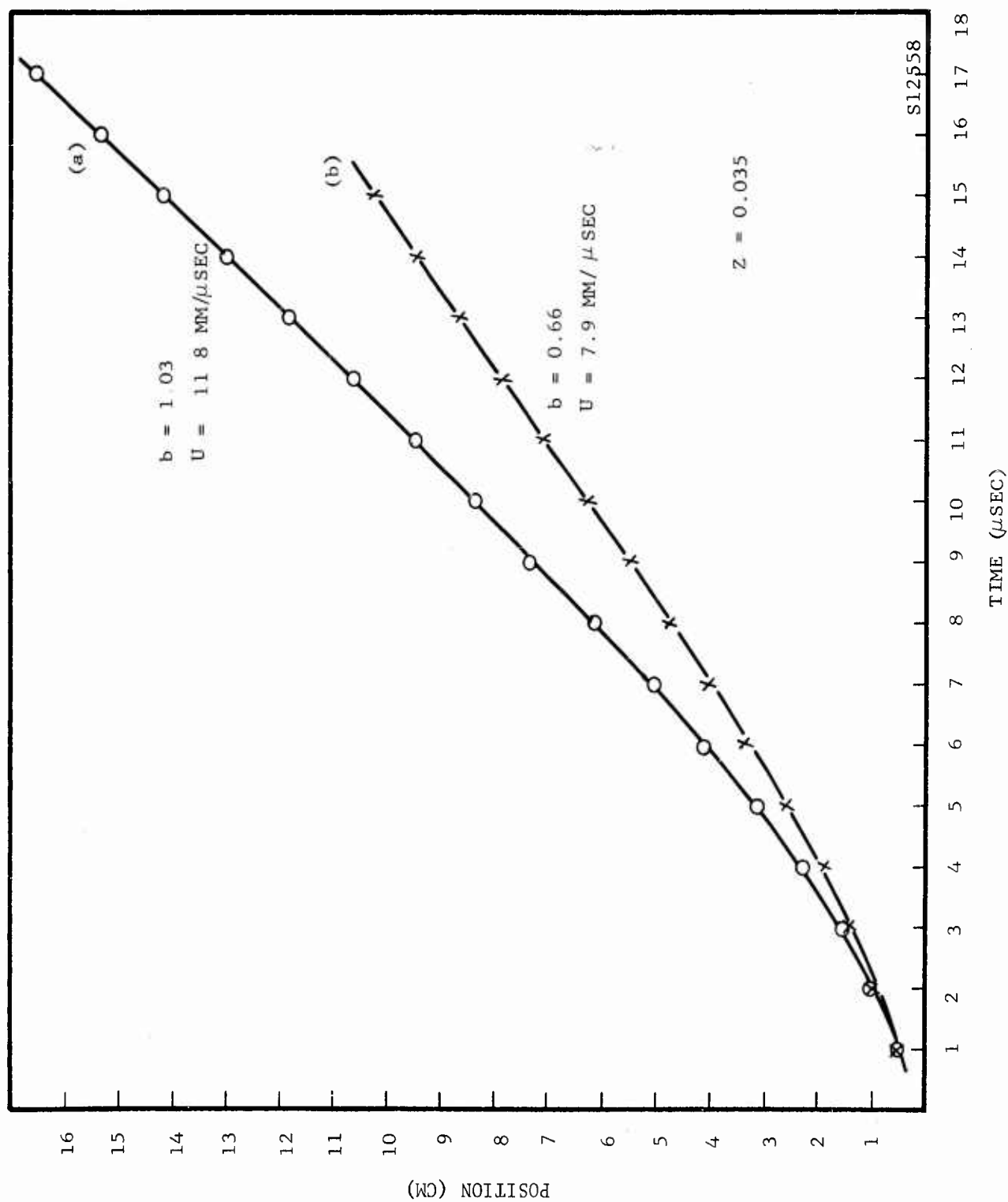


FIGURE 1. EFFECT OF PARAMETER b ON WAVE TRAJECTORY

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These were $b = 1.03$ (the value used in all previous calculations on this program) and $b = 0.66$. At the specific volumes appearing in the problems, these values of b correspond to maximum γ 's of 5.3 and 3.8, respectively.

Results of the calculation are shown on Figure 1, curves a and b. Other input data were as indicated in the previous section for curve "a". Use of $b = 0.66$ leads to a steady state velocity of 7.9 mm./ μ sec, much closer to the value of about 7 mm./ μ sec deduced from the theoretical steady state treatment than the 11.8 mm./ μ sec shown for $b = 1.03$. It is clear that a slight additional adjustment in "b" would produce the correct behavior.

The value of $b = 1.03$ was deduced, as discussed in Reference 2 from some data on the equation of state of explosive reaction products obtained by M. A. Cook. This data, and in particular its use for the equation of state of the solid phase is not beyond question. Nevertheless, before attributing the high steady state velocities to uncertainty in this data or its application to the present problem, it was thought desirable to re-examine the mathematical procedures for other possible sources of error. It has appeared that the steady state velocities may depend in a very significant way upon the details of the numerical integration in the vicinity of the shock front, and that methods now used may be inadequate in this region.

The integration presently makes use of a procedure devised by von Neuman and Richtmeyer⁴ for integrating numerically through the mathematical discontinuity which exists at a shock. Their procedure was to add a quantity "q" to the equations of motion. The "q" is generally likened to an artificial viscosity and is so constituted that it converts the shock discontinuity into a continuous function which changes rapidly over a few space zones from the situation in front of the shock to the situation behind the shock. It is designed to produce the correct shock energy, velocity, and pressure relations before and after the regions in which it is active, but falsifies these variables within this region. It was shown also to provide for the correct work loss due to entropy increase across the shock region.

The so-called von Neuman "q" was originally used with unreactive shocks and provides for the correct treatment of such shocks by numerical methods. In the case of reactive shocks such as occur in detonation, it can again be shown that the use of the "q" in the conservation equations does not affect the energy, pressure and velocity quantities on the two sides of the front. However, in this case, definition of a propagation velocity requires an additional relation generally known as the Chapman-Jouguet condition. It now appears that under some circumstances use of the von Neuman "q" can lead to an incorrect Chapman-Jouguet condition. This point can be understood by means of the following arguments.

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Figure 2a shows an actual pressure profile through a detonation wave as envisioned in terms of the hydrodynamic theory. The wave is initiated as a shock in which the pressure rises discontinuously from p_0 to p_2 . Reactions start at pressure p_2 and continue until completion at the Chapman-Jouguet point, p_1 .

Figure 2b shows the same wave computed using equations which include the von Neuman "q". Here the wave initiates with a gradually rising continuous pressure front which has a rounded off peak pressure, p_2 . In between there is a region of steeply rising pressure. The "q" used in the calculation is computed continuously through the rising pressure region as proportional to the square of the rate of compression, and provides for the same total energy deposition at the peak of the wave, p_2 , as in a shock process. In both cases the propagation velocity is given by

$$U = \frac{1}{p_0} \sqrt{-\frac{p_1 - p_0}{v_1 - v_0}} \quad (1)$$

With the situation represented as in Figure 2b, it is evident that if chemical reaction in the charge is readily initiated, such initiation may take place during the initial part of the pressure rise and reaction may be largely complete by the time the material has passed through the front and reached the point, p_2 . Thus, in contrast to the situation represented in Figure 2a, a major part of the chemical reaction and the corresponding heat release occurs, in effect, as part of the wave shock process. This has been observed to occur in typical examples of computed waves.

The consequences of this behavior in terms of wave velocity can be appreciated by consideration of a typical Hugoniot diagram as shown on Figure 3. According to the conventional interpretation^{5,6}, Figure 2a is represented on such a diagram as a process in which an increment of material starts at 0 as undisturbed material, jumps to 2 on passing through the lead shock and then moves down the line 21 while undergoing reaction, to 1, the Chapman-Jouguet point. At point 2, energy deposited by the shock in unreacted material is given by area 042. In the subsequent expansion to point 1, work represented by area 4216 is performed and the reaction energy, Q , is released. Thus, at point 1, energy remaining in the system is given by area 061 + Q . In the representation of the process shown in Figure 2b on a Hugoniot diagram, it is convenient to consider the extreme case in which reaction has gone to completion by the time point 2 is reached. In this circumstance, points 1 and 2 coincide and on Figure 3 the system goes directly from point 0 to point 1. Since this is treated as a shock transition, energy equivalent to area 061 is deposited in the process. The system energy at point 1 therefore is equivalent to area 061 + Q , exactly as before.

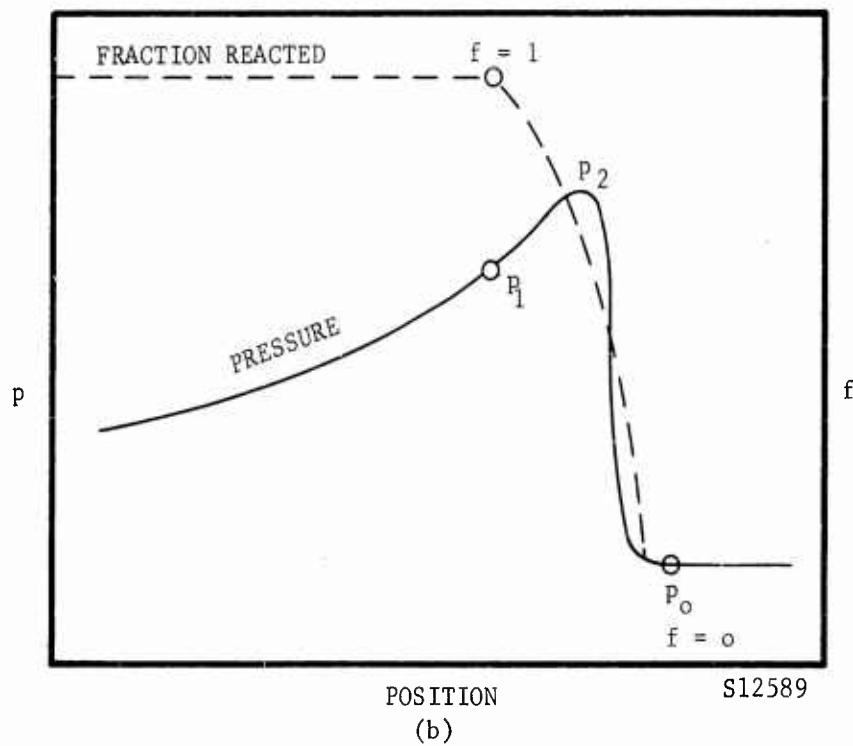
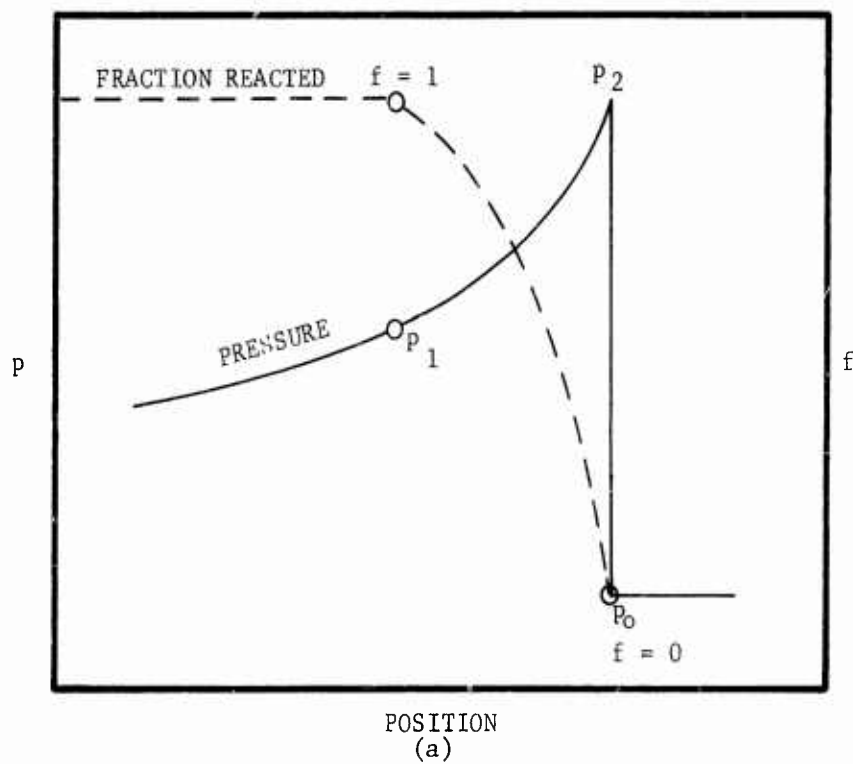


FIGURE 2. WAVE PROFILES WITH AND WITHOUT VON NEUMAN Q



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The first process can be considered as an isentropic compression along the adiabat 053, followed by heating (via shock dissipation) along 32 with a corresponding increase in entropy given by

$$\int_3^2 TdS = \text{area } 032, \text{ thence isentropic expansion along the adiabat } 27,$$

followed by heating due to reaction along 71, the latter involving a

$$\text{further entropy increase given by } \int_7^1 TdS = Q \text{ (the heat of reaction).}$$

The second process is made up of compression along the adiabat 05, and heating along 51 (due to chemical reaction and shock dissipation)

$$\text{with entropy increase corresponding to } \int_5^1 TdS = \text{area } 051. \text{ Considering}$$

that temperatures are highest at the Chapman-Jouguet point, it is apparent from these relations that the entropy increase in the second process is smaller than the first. Since the final energy content is the same in both cases, it must be concluded that the second process results in a higher pressure at the Chapman-Jouguet point than does the first. The second process cannot, therefore, terminate at point 1, but must instead arrive at some point at a higher pressure, such as 8. From equation (1), the relative propagation velocities in the two cases will be given by ratio of the square root of the slopes of the lines 01 and 08. It thus becomes evident that the situation represented by Figure 2b will correspond to a higher velocity than the value deduced from the steady state treatment.

The foregoing considerations further suggest that propagation velocities as deduced from the steady state treatment will be obtained if reaction is not permitted to occur until the system has passed completely through the shock zone, i.e., until point p_2 , Figure 2b, or point 2, Figure 3, is reached. In effect, this forces the system to go through the sequence, shock compression from point 0 to 1 (Figure 3) and expansion with reaction along line 21, as is implied in the steady state treatment. This has been subjected to verification by computing a wave trajectory in which the IBM 709 was arbitrarily programmed to provide such a reaction delay. Results are shown on Figure 4 as curve 1. For comparison purposes, curve 2 is included which is the trajectory shown on Figure 5 of the last quarterly report², obtained with no restrictions on reaction. Other input data was identical for the two curves and is as previously reported².

The difference between the two curves is pronounced, and indicates that the effect discussed is very important. Because of the long reaction zone existing in the wave, curve 1 never accelerated to steady

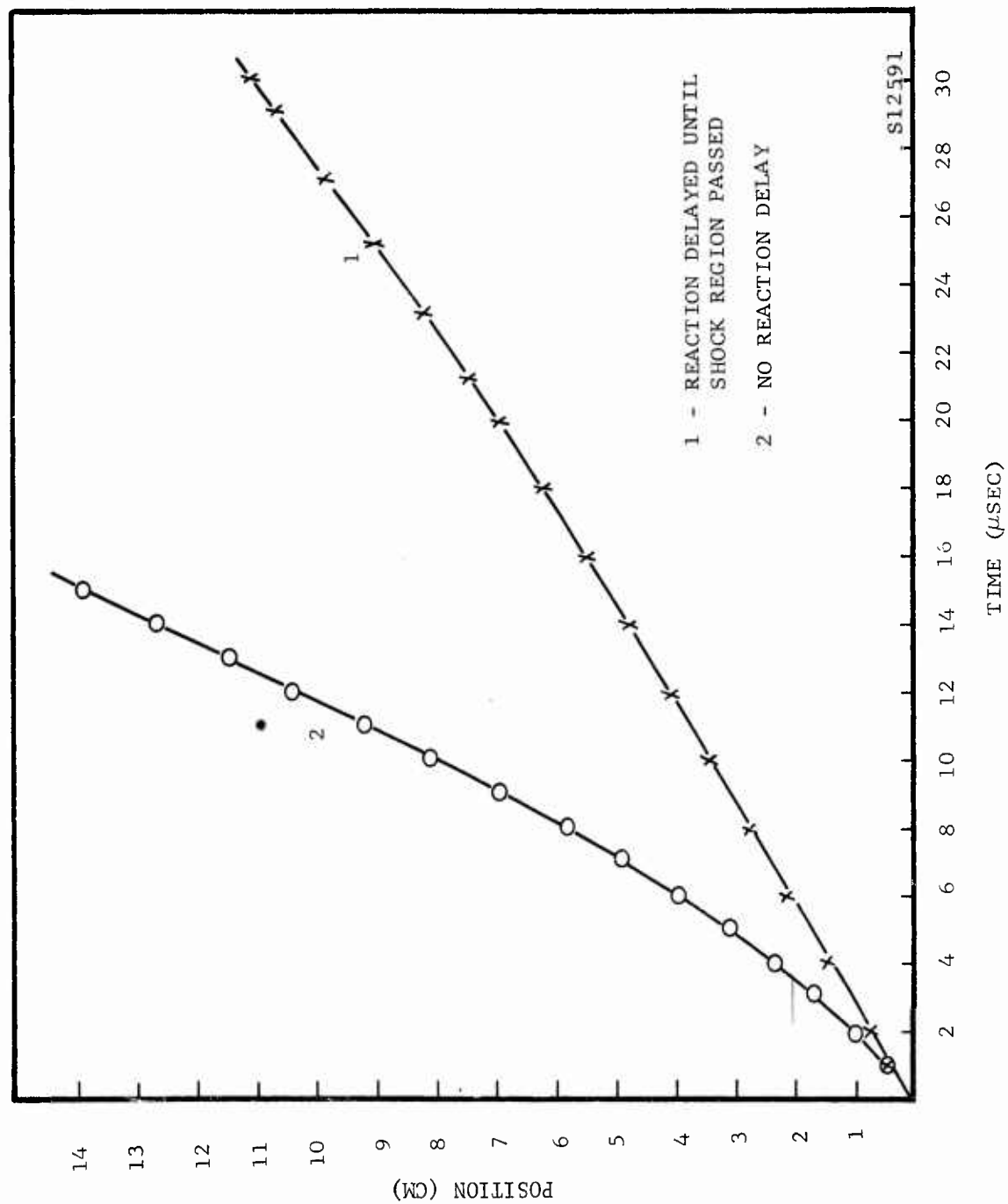


FIGURE 4. WAVE TRAJECTORIES SHOWING EFFECT OF PROGRAMMED REACTION DELAY

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state. It is therefore not ascertained whether the programmed reaction delay leads to the correct steady state velocity. However, it is clear that the details of the mathematical treatment in the vicinity of the wave shock zone are more important than has heretofore been thought to be the case. A number of procedures for obtaining a more correct treatment are possible and these are presently being considered.

It should be noted that this problem does not affect the validity of the calculations on minimum pressure for initiation of explosive materials, as discussed in previous reports^{2,3}. This is because in those cases, the critical point was that at which reaction ceased in the wave. At this point, the wave becomes an unreactive shock, and the present procedures are correct for an unreactive shock.

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SECTION 3

TWO DIMENSIONAL PROGRAM

Mathematics and coding are essentially complete and debugging is now in process on this program. However, it is subject to the considerations discussed in the previous section and may have to be delayed until improved procedures for treating the shock transition are developed. Any such delay should be of short duration. Every effort is being made to get this program running as soon as possible.

SECTION 4

EQUATION OF STATE STUDIES

The equation of state studies have involved some work on the refinement of techniques and calibration of equipment. However, a major part of the effort during this report period was directed to studies on the equation of state of polyurethane.

Methods were as described in the last quarterly report. Data is obtained in the form of a plot of pressure vs. piston movement over the up and down legs of a pressurization cycle. These legs diverge from the ambient pressure point due to a combination of piston sliding friction, and sample internal friction. This is the same in both directions⁷ and therefore symmetrical around a center curve except for a distorted region at each end due to the reversal of motion.

The magnitude of the frictional forces is indicated by the horizontal separation of the up and down legs in the symmetrical region. Press sliding friction is a function of pressure but independent of specimen material so that variations in separation for different materials or conditions are normally due to differences in sample internal friction, i.e., viscosity. Information on this property is of significant scientific interest, particularly in the present instance since it may afford some insight into possible differences between the equation of state observed under the present static methods, and that which applies to conditions of shock loading as encountered in a detonation process. No attempt at a detailed analysis on this basis has been made. The question will be examined further at a later date.

Data* on two typical press cycles to 26 and 40 kilobars and at ambient temperature are shown in Figure 5 as plots of $\Delta l/l$ vs. pressure (l = sample length). The values of Δl include a correction for press

*Data on the polyurethane specimen are shown in Table I.

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TABLE I
PREPARATION OF POLYURETHANE SPECIMEN

Polymer

Polypropylene glycol 2025	83.30%
Trimethylol propane	3.19%
Ferric acetylacetonate	0.125%
Toluene - 2,4 - diisocyanate	13.85%
Cure	24 hours @ 160 F.

Sample

Density	1.068 g/cm ³
Sample wt.	5.0532 g
Sample initial volume	4.729 cm ³

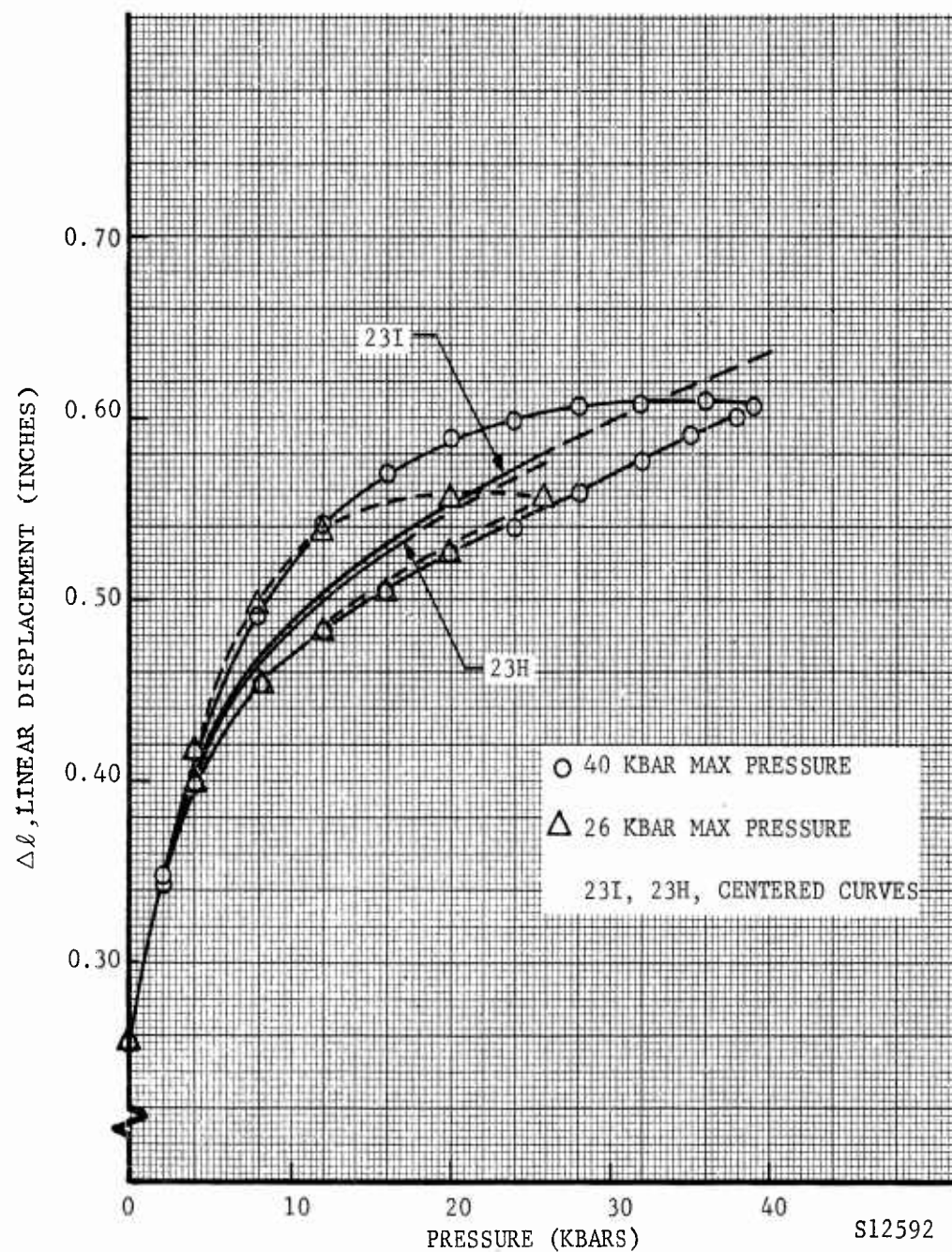


FIGURE 5. PRESS CYCLES FOR POLYURETHANE TEMP. - 23.3°C 0.5 INCH CYLINDER

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distortion obtained by running a similar cycle with no specimen in the pressure chamber. The midpoint between the up and down legs is indicated as curves 23I and 23H which are extrapolated (dashed portion) beyond the region of symmetry. The validity of the symmetry assumption is supported by the close agreement of these centered curves.

The press cycle and centered curve for polyurethane at 100°C is shown in Figure 6. Evident here is an increase in compressibility and a decrease in internal friction, the latter shown by the reduced separation between the up and down legs. Increase in compressibility with temperature is normal behavior for many organic substances.

Final p-v curves for the ambient and 100° temperatures are shown in Figure 7. A slight increase in volume as well as the increase in compressibility with temperature is noted at ambient pressure. Because of this, the curves for the two temperatures cross at about 2.5 kbar. In the region of 25 to 40 kbar the compressibility decreases with temperature as would be expected.

Attempts at interpretation of this work in terms of a general equation of state will be deferred until more complete data becomes available.

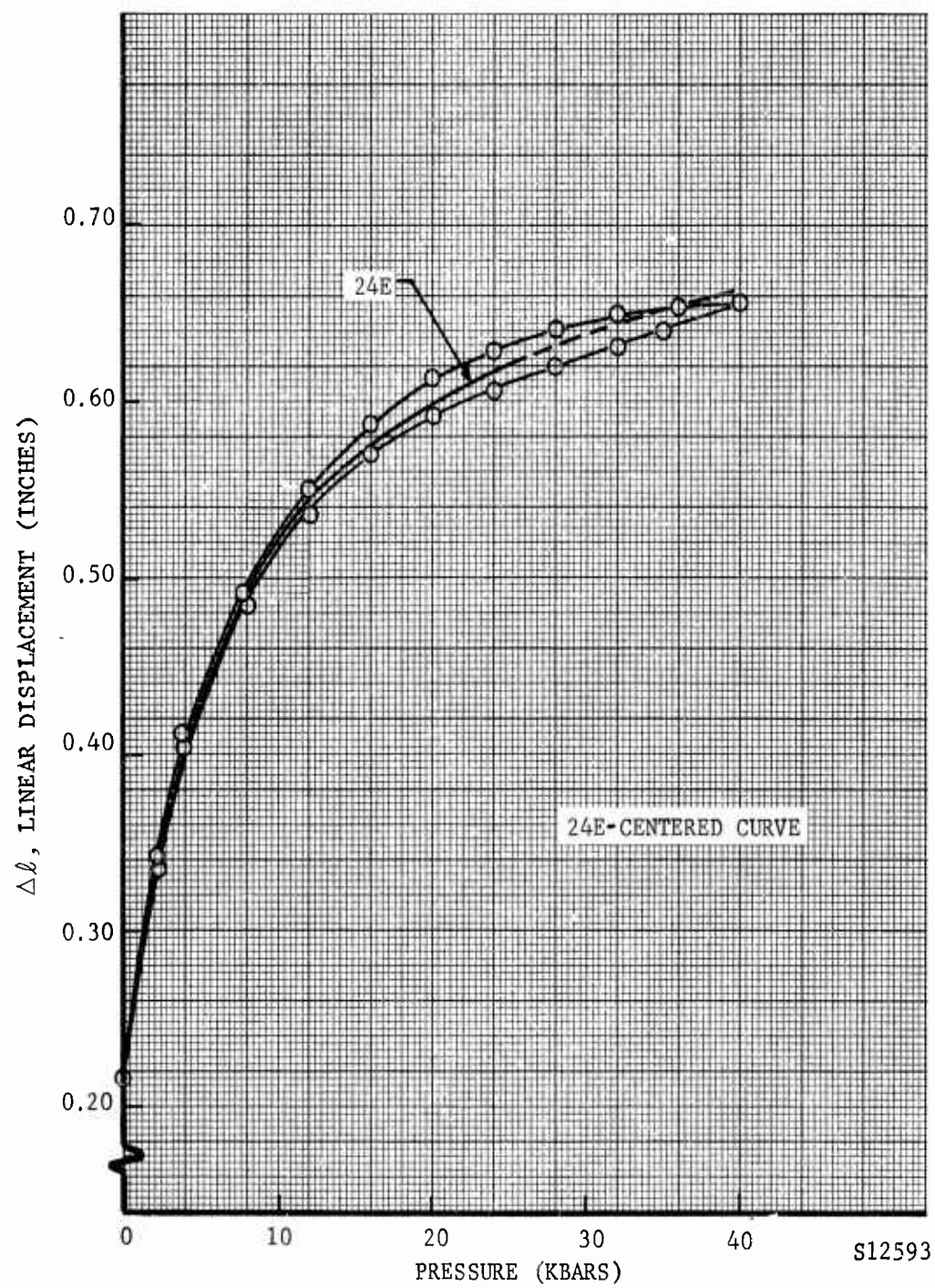


FIGURE 6. PRESS CYCLE FOR POLYURETHANE. MAXIMUM PRESSURE - 40 KBAR, TEMP. RANGE 99.5 - 101°C 0.5 INCH CYLINDER.

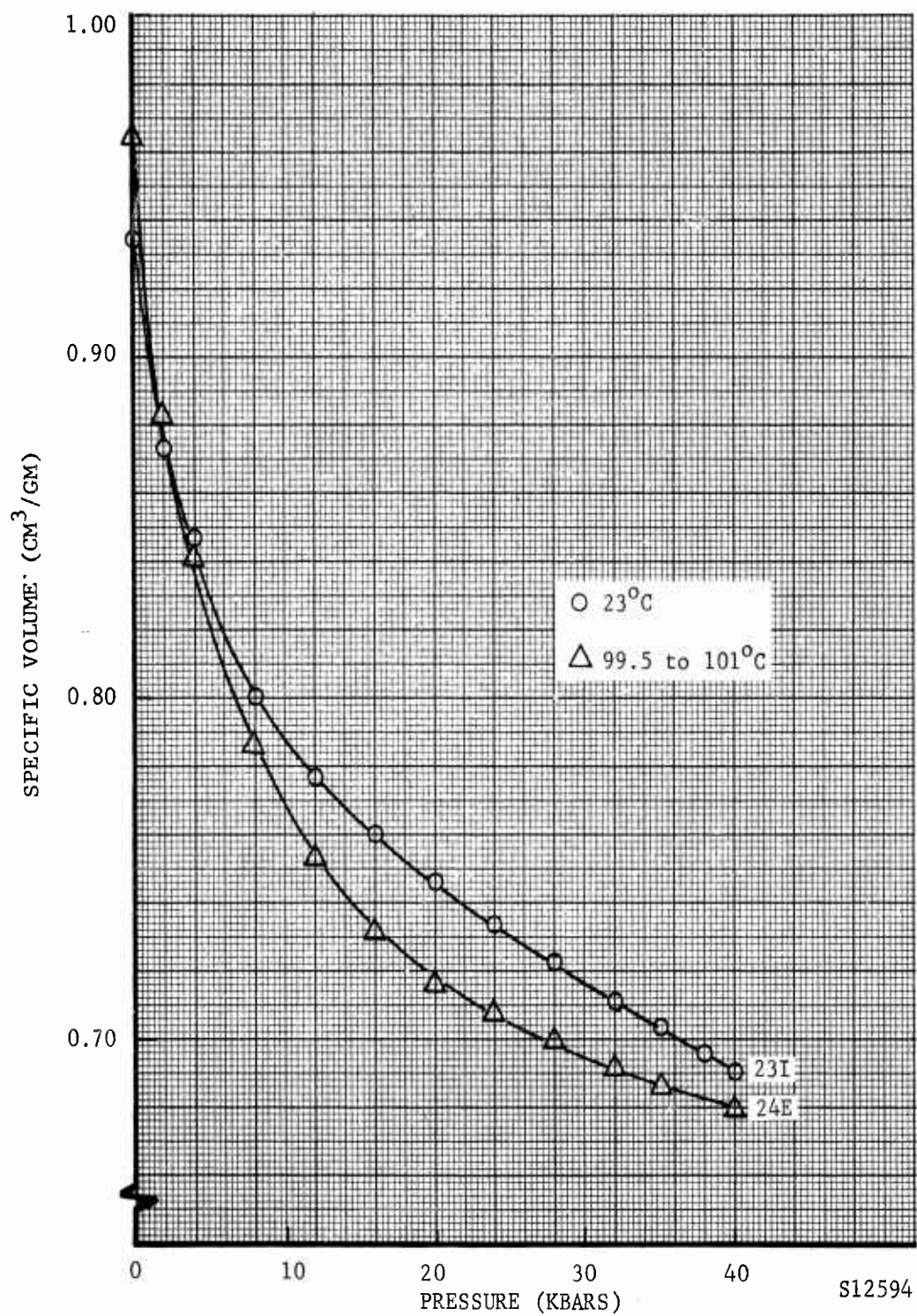


FIGURE 7. PRESSURE - VOLUME CURVES FOR POLYURETHANE

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